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REPLY TO "COMMENTS ON PAPERS BY OLANDER REGARDING ACTINIDE REDISTRIBUTION IN MIXED-OXIDE FUELS" BY R. O. MEYER

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Reply to "Comments on Papers by Olander Regarding Actinide Redistribution in Mixed-Oxide Fuels" by R. O. Meyer

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Meyer has advanced three criticisms of the models of actinide redistribution presented in references 1-3 of his note. The first of Meyer's criticisms is incorrect and the last two are nitpicking.

Conservation Equations

Meyer's criticism of the plutonium redistribution analysis presented in Ref. 1 of his note concerns the method of describing the mass flux of plutonium. The method of computing both the total mass flux and the mass flux of plutonium is based upon Fig. 1 of Meyer's note. In essence, a pore of volume V_p originally one one side of a test plane in the fuel is exchanged with an equal volume of solid originally on the other side of the test plane. The mechanism by which the solid is displaced from one side of the plane to the other is immaterial. It could have been moved by vapor transport, by ~~solid~~ surface diffusion, or, for that matter, by an ice cream scoop. Meyer agrees that the flux of the heavy metals is appropriately described by:

$$\text{mass flux of U and Pu} = -\rho_s V_p J_p + \rho_v V_p J_p \qquad (1)$$

Inasmuch as the solid and vapor phases consist of two components, it seems transparently obvious that the total mass flux can be broken down into two components:

$$\text{mass flux of U} = -\rho_s^U V_p J_p + \rho_v^U V_p J_p \tag{2a}$$

$$\text{mass flux of Pu} = -\rho_s^{Pu} V_p J_p + \rho_v^{Pu} V_p J_p \tag{2b}$$

where $\rho_s^U = (1-q)\rho_s$ and $\rho_s^{Pu} = q\rho_s$ are the masses of U and Pu per unit volume of solid, respectively, and $\rho_v^U = (1-u)\rho_v$ and $\rho_v^{Pu} = u\rho_v$ are the analogous mass densities in the vapor. The sum of Eqs (2a) and (2b) yields Eq (1), and Eq (2b) is equivalent to Eq (2) of Meyer's note.

Meyer derives the total mass flux expression by considering the consequences of exchanging a pore and the same volume of solid across the test plane. The correct plutonium flux formula can be derived simply by inserting the word "plutonium" at judicious points in the two sentences preceding his Eq(3)(the italics are added): "Movement of the pore across the test area has resulted in a net displacement of plutonium mass through the test area equivalent to the mass of plutonium in a solid sphere of volume V_p less the mass of plutonium in a vapor-filled sphere of the same volume V_p. Thus in this example, the total mass of plutonium transported is $q\rho_s V_p - u\rho_v V_p$, where ρ_s is the theoretical density of the solid phase, q is the mass fraction of plutonium in the solid and u is the mass fraction of plutonium in the vapor. Generalizing this, the plutonium mass flux expression is seen to be..." And Eq(2b) above follows.

Meyer's justification of his Eq(5) is based upon the sentence: "The diffusion flux of either species in the vapor phase is determined principally by the product of its diffusion coefficient and its vapor pressure.. This sentence amounts to a repeal of Fick's first law($J = -D(dc/dx)$) and replacing it by $J = Dc$, which of course has no basis. The diffusion flux of a species in a binary mixture is determined by the product of the diffusion coefficient and the vapor pressure gradient, not the vapor pressure proper. One simply cannot multiply the total mass flux by the mass fraction of one species in order to obtain the mass flux of this species. This procedure is correct only for the convective portion of the species flux (which is not significant in the present discussion); it certainly does not apply to the diffusive portion of the species flux. Meyer's Eq(5), which is based upon this procedure, is simply incorrect.

There are many simplifications in the treatment of vapor migration in cracks (reference 2 of Meyer's note), but the one singled out by Meyer is the least consequential of all. His contention that Eq (7) of his note is the correct conservation statement for gas phase transport of UO_3 is unassailable. The use of the quasi-stationary approximation (his Eq 6)) is justified quantitatively in Section 3.3 of the original paper. Meyer appears to believe that quasi-stationary approximations to partial differential equations of the diffusion or heat conduction type are in principle incorrect. Application of the quasi-stationary state approximation to diffusion in the vapor transport calculation is entirely analogous to the use of the steady state form of the heat conduction equation in the computation of the radial temperature distribution in the fuel rod. In the latter case, the fuel surface temperature, the thermal conductivity and the volumetric heat source term are all changing with time due to restructuring. The thermal diffusivity in oxide fuel is $\sim 0.02\ cm^2/sec$, which is an order of magnitude smaller than the gas diffusivity of UO_3. The characteristic distance over which heat or matter is transported is the fuel pin radius in both cases. Therefore the characteristic diffusion time ($\sim$ radius^2/diffusivity) is longer for heat conduction than for vapor diffusion. This means that more time is required for the temperature distribution in the rod to respond to changes in the parameters in the differential equation or in the boundary conditions than is required for similar changes in the UO_3 diffusion equation or its boundary conditions. In either case, the characteristic heat or mass diffusion times are on the order of seconds while the boundary conditions or coefficients in the conservation equations change over periods of hours or days. Consequently, if the quasi-stationary approximation is adequate for thermal analysis of the fuel pin, it certainly is acceptable for gas transport in cracks.

Vapor Migration in Pores

Meyer points out that modeling a pore as an infinite slab of vapor precludes leakage of plutonium around the edges, as would occur in a real pore. The parameter which determines the validity of the one-dimensional approximation for a disk-shaped pore is the ratio of the ~~pore~~ disk diameter, d, to the penetration distance of the plutonium concentration distribution attached to the leading ~~edge~~ face of the ~~pore~~ disk. In reference 3 of Meyer's note, the latter is shown to be $\sim D_s/v_p$, where D_s is the mutual diffusion coefficient of the heavy metal cations in the mixed oxide solid and v_p is the velocity of the migrating pore. The diameters of lenticular pores are typically 10 times greater than the pore thickness, or $d \sim 100 - 200$ μm. At 2000°K, D_s/v_p was found to be ~ 1μm. One would expect that "edge effects" would be felt to radial distances from the pore edge as much as several units of (D_s/v_p), or to several μm. Were this the case, 92% of the pore surface would behave as if it were part of an infinite pore and the outer annulus containing 8% of the surface would feel the finiteness of the pore. Neglecting radial diffusion (and hence the edge effect) in the solid on the leading edge of the pore when $d/(D_s/v_p) \simeq 10^2$ is probably as justifiable as neglecting axial heat conduction in a fast reactor fuel pin thermal analysis wherein the fuel length-to-diameter ratio is ~ 120.

Conclusion

The conclusion of study of actinide redistribution by migrating pores was <u>not</u> that the latter is incapable of segregating the former. This conclusion follows only if the ratio (D_s/v_p) is zero.

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